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International Bureau

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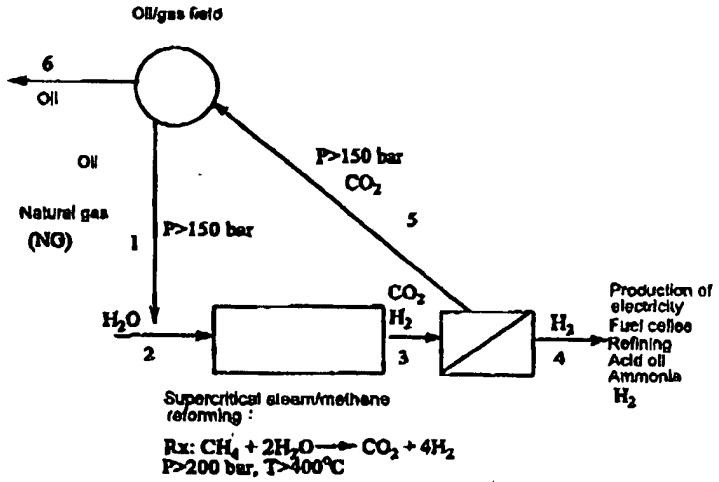
Published

With international search report.

Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.

In English translation (filed in Norwegian).

(54) Title: METHOD FOR PREPARING A H_2–RICH GAS AND A CO_2–RICH GAS AT HIGH PRESSURE



(57) Abstract

The present invention concerns a method for preparing a CO_2–rich gas stream for injection purposes or deposition, and a hydrogen rich gas stream, the method comprising the following steps: a) natural gas and H_2O are fed into a one–step reforming process for preparing a gas mixture comprising CO_2 and H_2 under supercritical condition for water from about 400 °C to about 600 °C, andpressure from about 200 to about 500 bar in the reforming reactor; b) the gas mixture from a) is separated into a H_2–rich and a CO_2–rich gas stream, respectively. The invention also comprises use of CO_2–rich gas stream for injection into marine formations, and use of H_2–rich gas stream for hydrogenation, as a source of energy/fuel in fuel cells and for production of electricity.

PATENT COOPERATION TREATY

BRYN & AARFLOT A/S

14 APR. 2000 PCT

From the INTERNATIONAL BUREAU

To:
BRYN & AARFLOT AS
P.O.Box 449 Sentrum
N-0104 Oslo
NORVEGE

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

Date of mailing (day/month/year) 06 April 2000 (06.04.00)	
Applicant's or agent's file reference 101529TFM	**IMPORTANT NOTICE**

International application No.	**International filing date** (day/month/year)	**Priority date** (day/month/year)
PCT/NO99/00283	15 September 1999 (15.09.99)	16 September 1998 (16.09.98)

Applicant
DEN NORSKE STATS OLJESELSKAP A.S et al

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:
   AU,CN,JP,KP,KR,US

   In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:
   AE,AL,AM,AP,AT,AZ,BA,BB,BG,BR,BY,CA,CH,CR,CU,CZ,DE,DK,DM,EA,EE,EP,ES,FI,GB,GD,GE,GH,GM,HR,HU,ID,IL,IN,IS,KE,KG,KZ,LC,LK,LR,LS,LT,LU,LV,MD,MG,MK,MN,MW,MX,NO,NZ,OA,PL,PT,RO,RU,SD,SE,SG,SI,SK,SL,TJ,TM,TR,TT,TZ,UA,UG,UZ,VN,YU,ZA,ZW
   The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on
   08 April 2000 (06.04.00) under No. WO 00/18681

REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, **a demand for international preliminary examination** must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the **national phase**, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO **34, chemin des Colombettes** **1211 Geneva 20, Switzerland**	**Authorized officer** J. Zahra
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38

Continuation of Form PCT/IB/308

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

Date of mailing (day/month/year)	IMPORTANT NOTICE
06 April 2000 (06.04.00)	
Applicant's or agent's file reference	**International application No.**
101529TFM	PCT/NO99/00283

The applicant is hereby notified that, at the time of establishment of this Notice, the time limit under Rule 46.1 for making amendments under Article 19 has not yet expired and the International Bureau had received neither such amendments nor a declaration that the applicant does not wish to make amendments.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/NO 99/00283

A. CLASSIFICATION OF SUBJECT MATTER

IPC7: C01B 3/48, C01B 31/20
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7: C01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

FULLTEXT, EPODOC, WPI

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CA 868821 A (GIORGIO PAGANI), 20 April 1971 (20.04.71), page 3, line 9 - line 12, claims --	1-6
X	WO 9829333 A1 (EXXON CHEMICAL PATENTS INC.), 9 July 1998 (09.07.98) --	1-6
X	US 5714132 A (AKHILESH KAPOOR ET AL), 3 February 1998 (03.02.98), column 1, line 11 - line 14; column 2, line 38 - line 44; column 5, line 21 - line 26 --	1-6
A	US 3652454 A (ALLEN M. ROBIN ET AL), 28 March 1972 (28.03.72) --	1-6

☐ Further documents are listed in the continuation of Box C. ☒ See patent family annex.

* Special categories of cited documents
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" erlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
10 March 2000	14 -03- 2000
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. +46 8 666 02 86	Authorized officer Helena Hemphälä/ELY Telephone No. +46 8 782 25 00

INTERNATIONAL SEARCH REPORT

International application No.
PCT/NO99/00283

Box I Observations where certain claims were found unsearchable (Continuation of item 1 f first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

See extra sheet.

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.: 1-. 6

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.
☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/NO99/00283

I. Claims 1-6 relate to a process for producing a CO2- and a H2-rich gas.

II. Claim 7 relates to the use of a CO2-rich gas for injection in marine formations.

III. Claim 8 relates to the use of a H2-rich gas for hydrogenation.

IV. Claim 9 relates to the use of a H2-rich gas as a source of energy in a fuel cell.

V. Claim 10 relates to the use of a H2-rich gas for electricity production.

The special technical feature of Group I is a process for producing the gases H2 and CO2. The special technical features of Group II-V involve different applications of the gases H2 and CO2. The gases H2 and CO2, produced according to claim 1, are well known products. There is a reference in each of the independent claims 7-10 to the process for producing these known products. However, the production process does not give the products any new characteristics, therefore the reference made in claim 7-10 does not involve the features in claim 1. The group of inventions is not so linked as to form a single inventive concept under PCT Rule 13.

INTERNATIONAL SEARCH REPORT

Information on patent family members

02/12/99

International application No.
PCT/NO 99/00283

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
CA 868821 A	20/04/71	NONE	
WO 9829333 A1	09/07/98	AU 6646498 A	31/07/98
		EP 0950017 A	20/10/99
		NO 993228 A	30/08/99
		US 5904880 A	18/05/99
US 5714132 A	03/02/98	EP 0643013 A	15/03/95
		ZA 9405891 A	13/06/95
US 3652454 A	28/03/72	NONE	

PU

PATENT COOPERATION TREATY

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

BRYN & AARFLOT A/S

To:

BRYN & AARFLOT AS
P.O.Box 449 Sentrum
N-0104 Oslo
NORVEGE

22 JUNI 2000

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT
(PCT Rule 71.1)

Date of mailing *(day/month/year)*	19.06.2000
Applicant's or agent's file reference	IMPORTANT NOTIFICATION
101529TFM	

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/NO99/00283	15/09/1999	16/09/1998

Applicant
DEN NORSKE STATS OLJESELSKAP A.S et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.

2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.

3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/	Authorized officer
European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Gregoire, J-P Tel.+49 89 2399-8041

Form PCT/IPEA/416 (July 1992)

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
101529TFM		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/NO99/00283	15/09/1999	16/09/1998
International Patent Classification (IPC) or national classification and IPC		
C01B3/32		
Applicant		
DEN NORSKE STATS OLJESELSKAP A.S et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 4 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
11/04/2000	19.06.2000
Name and mailing address of the international preliminary examining authority:	Authorized officer
European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Van Iddekinge, R Telephone No. +49 89 2399 8346

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/NO99/00283

I. Basis f the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

Description, pages:

1-6 as published

Claims, No.:

1-10 as published

Drawings, sheets:

1 as published

2. The amendments have resulted in the cancellation of:

☐ the description, pages:
☐ the claims, Nos.:
☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

☐ the entire international application.

☒ claims Nos. 7-10.

because:

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/NO99/00283

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

☐ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. are so unclear that no meaningful opinion could be formed (*specify*):

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

☒ no international search report has been established for the said claims Nos. 7-10.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-6
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-6
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-6
	No:	Claims	

2. Citations and explanations

see separate sheet

INTERNATIONAL PRELIMINARY EXAMINATION REPORT - SEPARATE SHEET

International application No. PCT/NO99/00283

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1). An international search report has been made for claims 1-6. Claims 7-10 have not been searched, because of lack of unity. Therefore no substantive examination is possible for claims 7-10.

Re Item V

Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

2). Reference is made to the following documents:
D1=CA-A-868821
D2=WO-A-9829333
D3=US-A-5714132

Novelty

3). None of the available documents of the search report including D1, D2 and D3 disclose step a of claim 1.

Therefore claims 1 and its dependent claims 2-6 fulfil the requirements of Article 33(2) PCT (novelty).

D1 and D3 are not relevant for the inventive step, because they are concerned with the production of hydrogen and carbon monoxide. D2 is concerned with the production of hydrogen and carbon dioxide, but it is not relevant because a different feed is used (methanol instead of natural gas).

Therefore claims 1 and its dependent claims 2-6 fulfil the requirements of Article 33(3) PCT (inventive step).

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Method for preparing a H_2-rich gas and a CO_2-rich gas at high pressure.

The present invention comprises a method for production of a CO_2-rich gas for injection purposes or to be deposited, and a hydrogen-rich gas, and use thereof.

It is commonly assumed that the greenhouse effect and the climate on earth have are closely connected to human made emissions of CO_2. These emissions are primarily formed by combustion of coal and hydrocarbons, i.a. by generation of heat and electric power. A desireable goal is therefore to reduce the emission of CO_2 to the atmosphere.

It is known art to reduce the emission of CO_2 from combustion of natural gas, e.g. by gas reforming and shift technology for preparation of a mixture consisting of hydrogen and carbon dioxide. These components are then separated, whereafter hydrogen is used as fuel in a gas turbin and carbon dioxide is deposited after compression to desired pressure. The deposition can be made on the bottom of the sea or in geological reservoires. The reservoires can also contain hydrocarbons. The above mentioned technique is i.a. described in Teknisk Ukeblad No. 16, page 8, 1998.

Known art comprising gas reforming and shift technology as described above is very expensive and at the same time gives less energy yield than a conventional, modern gas power plant.

US 3,652,454 describes preparation of CO_2 and H_2 from a gas stream containing CO by an improved continuous catalytical shift reaction at high pressure. The reaction takes place i one or more shift reactors at a superatmospheric pressure of from 35 to 250 atmospheres, and a temperature between 287°C and 537°C. The patent does not describe reforming of natural gas.

From EP 0 000 993-A1 it is known a method for preparation of ammonia by means of a primary and a secondary catalytic reforming of an hydrocarbon stream at superatmospheric pressure. From the primary catalytic reforming the ratio of steam to carbon is from 2.5 to 3.5, the pressure is from 30 to 120 bar and the temperature out of the reactor is from 750 to 850°C. From the secondary catalytic reforming the content of methane is from 0.2 to 10 % by weight on a dry

basis and the ratio of hydrogen to nitrogen is from 2.2 to 2.7. To the he secondary reforming there is added an excess of air for preparing a gas with a higher content of methane, i.e. at a lower temperature, and/or a lower steam ratio and/or a higher pressure . In the above mentioned EP patent CO_2 is removed at a low pressure by taking out hydrogen at an elevated pressure for further use by the preparation of ammonia.

EP 0 289 419 describes catalytic steam reforming of hydrocarbons for preparing hydrogen i an ammonia process. The catalytic steam reforming takes place at a pressure from 25 to 120 bar, a temperature from 800 to 1000°C and at at ratio steam:carbon of 1.8-2.5. The process is operated in such a way that there are less than 0.3% impurities in the H_2-rich gas which is to be used for production of ammonia. The present invention allows a higher content than 0.3% of CO, CO_2 and CH_4 in the H_2-rich gas stream.

CA 868,821 describes preparation of synthesis gas by steam reforming of hydrocarbons in a gas and a liquid at 50-250 absolute atmospheres, preferably 160 abs. atm. for production of ammonia and methanol.

Known art does not deal with a one step prosess for production av CO_2-rich gas and H_2-rich gas under supercritical conditions for water, where a CO_2-rich gas mixture is taken out at an elevated pressure in the interval from 20 to 200 bar for injection or deposition i marine formations. The present invention involves reduced compression costs by deposition or injection in marine formations because the CO_2-rich gas mixture is taken out at an elevated pressure.

The present invention comprises a method for preparing a CO_2-rich gas stream and a hydrogen rich gas stream, the method comprising the following steps:

a) natural gas and H_2O are fed to a one-step reforming process for preparing a gas mixture comprising CO_2 and H_2 under supercritical conditons for water;

b) the gas mixture from a) is separated into a H_2-rich and a CO_2-rich gas stream, respectively.

Further the temperature in the reforming reactor is from about 400°C to about 600°C, and the method is also characterized by a pressure in the reform-

ing reactor from about 200 to about 500 bar. The CO_2-rich gas stream from the separation unit is at a pressure in the interval from 20 til 200 bar. I the present invention the mixture in the reforming reactor may be passed over a catalyst bed. The reforming can also be carried out without catalyst. The present invention also concerns use of the CO_2-rich gas stream prepared according to the previously mentioned method, where the CO_2-rich gas stream is injected into marine formations. Further, the invention comprises use of the H_2-rich gas stream prepared according to the invention, where the H_2-rich gas stream can be utilized for hydrogenation, i the production of electricity and as a source of energy / fuel in fuel cells.

The following reaction takes place during the reforming:

$$CH_4 + 2H_2O \rightarrow CO_2 + 4H_2$$

The reforming reactor is operated at supercritical conditions for water. The temperature in the reforming reactor is from about 400°C to about 600°C and the pressure in the reforming reactor is from about 200 to about 500 bar. It is an object of the present invention that CO_2 is separated from the gas stream at a pressure of at least 20 bar and maximum 200 bar before being injected into marine formations or by deposition. The reforming reaction takes place over a suitable catalyst bed. The reforming can also take place without catalyst in the reforming reactor. It is also an object of the present invention to use H_2 made according to the method of the invention, for hydrogation, and for production of electricity. Use of H_2 as a source of energy / fuel in fuel cells is further comprised by the present invention.

CO_2 is an acid gas, and the most widely used method to separate the mentioned gas from other non-acid gas molecules is absorption. During absorption the different chemical properties of the gas molecules are utilized. By contacting the gas mixture with a basic liquid the acid gases to a high degree will be dissolved in the liquid. The liquid is separated from the gas and the absorbed gas can the be set free either by altering the composition of the liquid or by altering pressure and temperature. For separation of CO_2 mainly aqueous solutions of alcoholamines are used. The absorption is taking place at a relatively low temperature and high pressure, while stripping of the gas from the liquid is carried

out at a relatively high temperature and low pressure. To liberate CO_2 from the amine phase in the stripping unit stripping steam is usually used. If the partial pressure of CO_2 in the gas into the absorber is high, e.g. higher than 15 bar, it is possible to obtain high concentrations in the amine phase, and a large part of absorbed CO_2 can be set free in the stripping unit at elevated pressure, e.g. 5-8 bar.

By the use of one or more semipermeable membrane units it is possible to achieve that molecules of different molecular weight and different properties permeate the membrane at different velocities. This principle can be utilized to separate gases. For the gas mixture in question membranes can be selected where H_2 permeates rapidly, whereas CO_2 permeates slowly, whereafter a separation-in-part of the different gas components is achieved. By combining solid membranes and liquid membranes it is also possible to achieve a rapid permeation of CO_2, while H_2 is kept back. It can be difficult to achieve complete separation of the different gas components by using different separation methods. This is especially the case by use of membranes. For gas mixtures which are going to be burned, a partly separation of hydrogen and CO_2 will be sufficient.

In the present invention it is desireable to deposit out-separated CO_2. Large amounts of CO_2 can be deposited according to various methods, of which the three most interesting are deposition at very deep oceans, deposition in deep water reservoirs and deposition in oil reservoirs wherin the gas at the same time functions as drive agent for enhanced oil recovery. The two last mentioned storage methods are operated commercially. In these storage forms the CO_2 gas has to be brought to high pressure for transport in pipelines to a deposition well and further to injection. The injection pressure will vary, but could be in the range 50 to 300 bar. If the CO_2 gas can be separated from the H_2/CO_2 mixture at an elevated pressure, significant compression work can be avoided, and this is the case in the present invention.

The invention is further elucidated on Figure 1. Natural gas (1) is passed from an oil/gas field, and blended with H_2O (2) before the mixture is passed to reforming at supercritical conditions. Produced synthesis gas (3) is separated at high pressure into two streams, a CO_2-rich stream (5), which is injected into an

oil/gas field, and a H_2-rich stream (4), respectively The H_2-rich stream is further used for hydrogenation, as a source of energy in fuel cells, and for production of electricity.

Example 1:

The example concerns one or more membrane units wherein the CO_2-rich gas can have a pressure approximately equal to the partial pressure of CO_2 into the separation unit, as shown in Table 1 below.

Table1

Total inlet pressure on the separation unit (bar)	Partial pressure of CO_2 out of the separation unit (bar)
200	40
250	50
300	60

Example 2:

Supercritical conditions occur at pressures above 220 bar and temperatures above 374°C.

In this example it is described at which conditions supercritical conditions occur in the present reactor.

The relationship between temperatur and pressure in the reactor in the present invention is as shown in Table 2. Supercritical conditions occur in the reactor when the values of pressure and temperatur are higher than shown in Table 2.

Table 2

Pressure (bar)	Temperature (°C)
1100	354
750	356
500	362
300	368
220	374

Relationship between temperature and % fraction H_20 at 220 bar is shown in table 3. Supercritical conditions occur in the reactor when the values for temperature and % fraction H_2O at a pressure of 220 bar are higher than shown in Table 3.

Table 3

% fraction H_2O (P =220 bar)	Temperature (°C)
0.95	372
0.85	365
0.75	353

The relationship between pressure and % fraction H_2O at a temperature of 374°C is shown in Table 4. Supercritical conditions occur in the reactor when the values for pressure and % fraction H_2O at 374°C are higher than shown in Table 4.

Table 4

% fraction H_2O (T= 374°C)	Pressure (bar)
0.95	300
0.85	400
0.75	1000

Claims

1. Method for preparing a CO_2-rich gas stream for injection purposes or deposition, and a hydrogen rich gas stream,
characterized in that the method comprises the following steps:

a) natural gas and H_2O are fed into to a one-step reforming process for preparing a gas mixture comprising CO_2 and H_2 under supercritical conditons for water;

b) the gas mixture from a) is separated into a H_2-rich and a CO_2-rich gas stream, respectively.

2. Method according to claim 1,
characterized in that the temperature in the reforming reactor is from about 400°C to about 600°C.

3. Method according to claims 1-2,
characterized in that the pressure in the reforming reactor is from about 200 to about 500 bar.

4. Method according to claims 1-3,
characterized in that the CO_2-rich gas stream is present at a pressure within the interval from 20 to 200 bar.

5. Method according to claims 1-4,
characterized in that the mixture in the reforming reactor is passed over a catalyst bed.

6. Method according to claims 1-5,
characterized in that the reaction in the reforming reactor is carried out without a catalyst.

7. Use of a CO_2-rich gas stream according to claim 1 for injection into marine formations.

8. Use of a H_2-rich gas stream made according to claim 1 for hydrogenation.

9. Use of a H_2-rich gas stream made according to claim 1 as a source of energy / fuel in fuel cells.

10. Use of a H_2-rich gas stream made according to claim 1 for production of electricity.

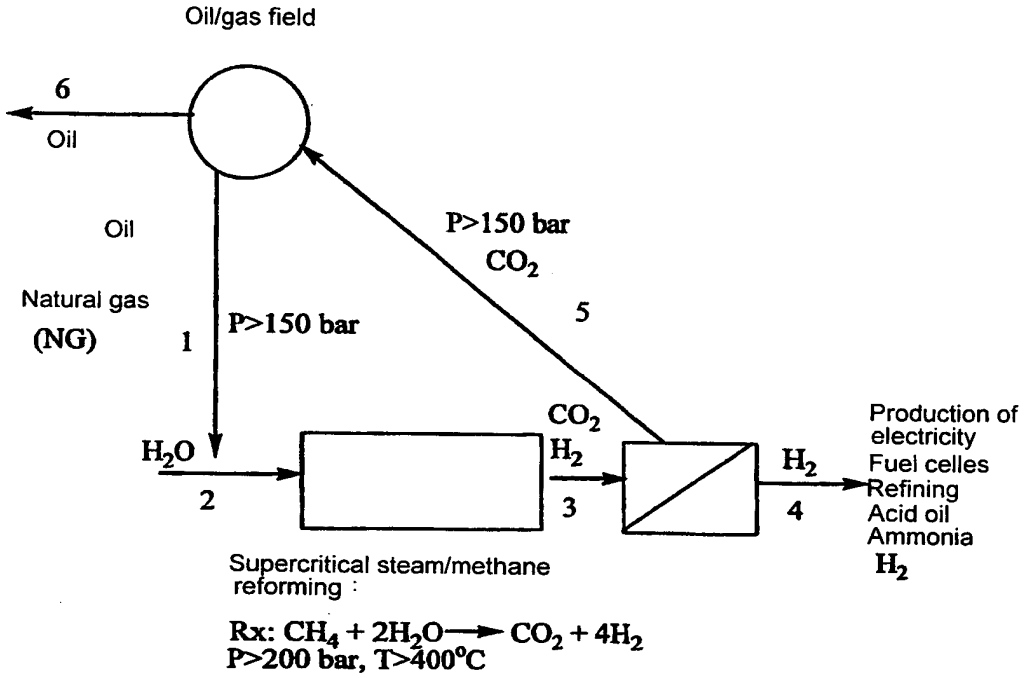
Oil/gas field
6
Oil
Oil
Natural gas
(NG)
1
P>150 bar
P>150 bar
CO_2
5
H_2O
2
CO_2
H_2
3
H_2
4
Production of electricity
Fuel celles
Refining
Acid oil
Ammonia
H_2
Supercritical steam/methane reforming :
Rx: $CH_4 + 2H_2O \rightarrow CO_2 + 4H_2$
P>200 bar, T>400°C

Figure 1

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PCT/NO99/00283

PCT REQUEST

101529TFM

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0 0-1	For rec iving Offic use nly Int rnational Application No.	PCT/NO 9 9 / 0 0 2 8 3
0-2	International Filing Date	15 SEPT. 1999 (15.09.99)
0-3	Name of receiving Office and "PCT International Application"	PATENTSTYRET Styret for det industrielle rettsvern PCT International application
0-4 0-4-1	Form - PCT/RO/101 PCT Request Prepared using	PCT-EASY Version 2.84 (updated 01.06.1999)
0-5	Petition The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty	
0-6	Receiving Office (specified by the applicant)	Norwegian Patent Office (RO/NO)
0-7	Applicant's or agent's file reference	101529TFM
I	Title of invention	PROCESS FOR PREPARING A H2-RICH GAS AND A CO2-RICH GAS BY HIGH PRESSURE.
II II-1 II-2 II-4 II-5 II-6 II-7	Applicant This person is: Applicant for Name Address: State of nationality State of residence	applicant only all designated States except US DEN NORSKE STATS OLJESELSKAP A.S N-4035 STAVANGER Norway NO NO
III-1 III-1-1 III-1-2 III-1-4 III-1-5 III-1-6 III-1-7	Applicant and/or inventor This person is: Applicant for Name (LAST, First) Address: State of nationality State of residence	applicant and inventor US only OLSVIK, Ola Haukv. 18 N-7562 Hundhammeren Norway NO NO

15.sep99 727852 PCT 99/00283 19 17.880,00

15.sep99 727853 PCT 99/00283 89 500,00

15.sep99 727854 PCT 99/00283 87 300,00

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IV-1	**Agent or common representative; or address for correspondence** The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:	agent
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IV-1-5	e-mail	email@baa.no
V	**Designation of States**	
V-1	Regional Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AP: GH GM KE LS MW SD SL SZ UG ZW and any other State which is a Contracting State of the Harare Protocol and of the PCT EA: AM AZ BY KG KZ MD RU TJ TM and any other State which is a Contracting Stat of the Eurasian Patent Convention and of the PCT EP: AT BE CH&LI CY DE DK ES FI FR GB GR IE IT LU MC NL PT SE and any other Stat which is a Contracting State of the European Patent Convention and of the PCT OA: BF BJ CF CG CI CM GA GN GW ML MR NE SN TD TG and any other State which is a member State of OAPI and a Contracting State of the PCT
V-2	National Patent (other kinds of protection or treatment, if any, are specified between parentheses after the designation(s) concerned)	AE AL AM AT AU AZ BA BB BG BR BY CA CH&LI CN CR CU CZ DE DK DM EE ES FI GB GD GE GH GM HR HU ID IL IN IS JP KE KG KP KR KZ LC LK LR LS LT LU LV MD MG MK MN MW MX NO NZ PL PT RO RU SD SE SG SI SK SL TJ TM TR TT UA UG US UZ VN YU ZA ZW
V-3	National Patent (States which have become party to the PCT after the issuance of this version of EASY)	TZ

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V-5	**Precauti nary D signati n Statement** In addition to th d signations made under items V-1, V-2 and V-3, th applicant also makes under Rule 4.9(b) all designations which would be permitted under the PCT except any designation(s) of the State(s) indicated under item V-6 below. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit.		
V-6	**Exclusion(s) from precautionary designations**	NONE	
VI-1	**Priority claim of earlier national application**		
VI-1-1	Filing date	16 September 1998 (16.09.1998)	
VI-1-2	Number	1998 4296	
VI-1-3	Country	NO	
VI-2	**Priority document request** The receiving Office is requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) identified above as item(s):	VI-1	
VII-1	**International Searching Authority Chosen**	Swedish Patent Office (ISA/SE)	
VII-2	**Request to use results of earlier search; reference to that search**		
VII-2-1	Date	11 June 1999 (11.06.1999)	
VII-2-2	Number	1998 4296	
VII-2-3	Country (or regional Office)	NO	
VIII	**Check list**	number of sheets	electronic file(s) attached
VIII-1	Request	4	-
VIII-2	Description	7	-
VIII-3	Claims	2	-
VIII-4	Abstract	1	101529abs.txt
VIII-5	Drawings	1	-
VIII-7	TOTAL	15	
	Accompanying items	paper document(s) attached	electronic file(s) attached
VIII-8	Fee calculation sheet	✓	-
VIII-11	Statement explaining lack of signature	✓	-
VIII-16	PCT-EASY diskette	-	diskette
VIII-17	Other (specified):	Copy of Official Action with Examination Report	-
VIII-18	**Figure of the drawings which should accompany the abstract**		
VIII-19	**Languag of filing of the internati nal application**	Norwegian	

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101529TFM

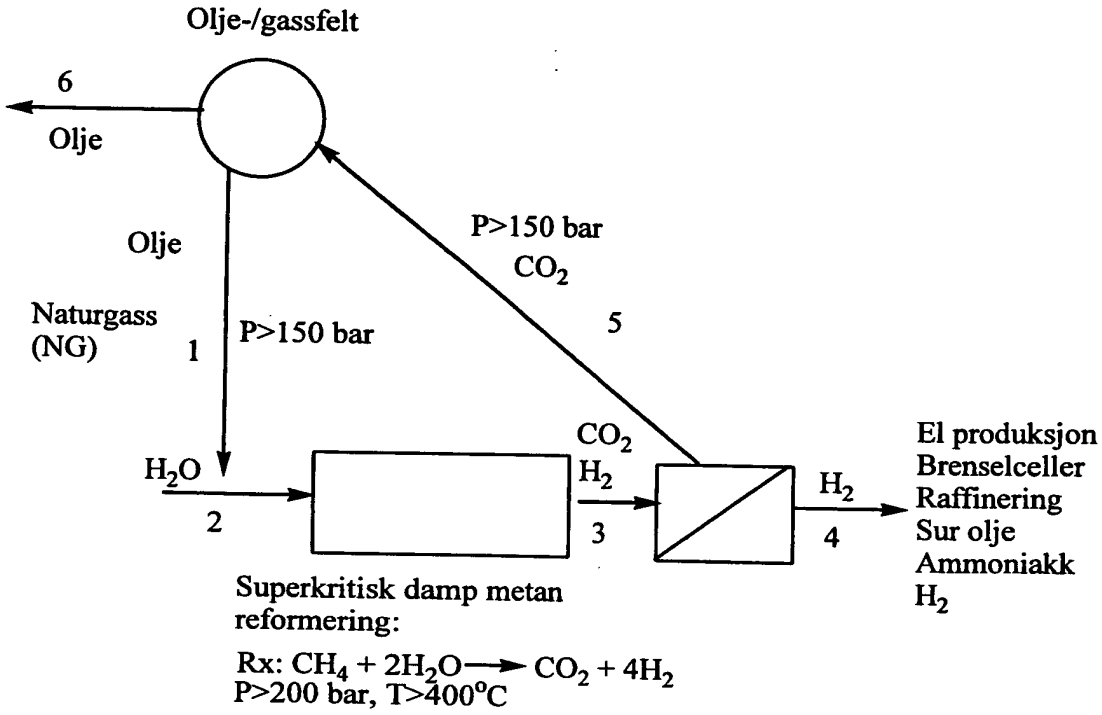
IX-1	Signature of applicant or ag nt	
IX-1-1	Name	BRYN & AARFLOT AS
IX-1-2	Name of signatory	MARKUSSEN, Tone Furan
IX-1-3	Capacity	Patent Attorney

FOR RECEIVING OFFICE USE ONLY

10-1	Date of actual receipt of the purported international application	15 SEPT. 1999 (15.09.99)
10-2	Drawings:	
10-2-1	Received	
10-2-2	Not received	Received
10-3	Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application	
10-4	Date of timely receipt of the required corrections under PCT Article 11(2)	
10-5	International Searching Authority	ISA/SE
10-6	Transmittal of search copy delayed until search fee is paid	

FOR INTERNATIONAL BUREAU USE ONLY

11-1	Date of receipt of the record copy by the International Bureau	30 SEPTEMBRE 1999 30.09.99



Figur 1

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Fremgangsmåte for å fremstille en H_2-rik gass og en CO_2-rik gass ved høyt trykk.

Foreliggende oppfinnelse omhandler en fremgangsmåte for å fremstille en CO_2-rik gass til injeksjonsformål eller deponering og en hydrogenrik gass samt anvendelse derav.

Det er alminnelig antatt at drivhuseffekten og klimaet på jorden har en nær sammenheng med menneskeskapt CO_2-utslipp. Dette utslippet dannes først og fremst ved forbrenning av kull og hydrokarboner, bl.a. ved generering av varme og elektrisk kraft. Et ønskelig mål er derfor å redusere utslippet av CO_2 til atmosfæren.

Det er tidligere kjent å redusere utslippet av CO_2 ved forbrenning av naturgass f.eks. ved gassreformerings- og skiftteknologi for fremstilling av en blanding bestående av hydrogen og karbondioksid. Videre separeres disse komponentene, hvoretter hydrogen anvendes som brennstoff i en gassturbin og karbondioksid deponeres etter komprimering til ønsket trykk. Deponeringen kan finne sted på havbunnen eller i geologiske reservoarer. Reservoarene kan også inneholde hydrokarboner. Ovennevnte teknikk er bl.a. omtalt i Tekniks Ukeblad nr. 16, side 8, 1998.

Kjent teknikk omfattende gassreformerings- og skiftteknologi som beskrevet ovenfor er særdeles kostbar og gir samtidig mindre energiutbytte enn et konvensjonelt, men moderne, gasskraftverk.

US 3,652,454 beskriver fremstilling av CO_2 og H_2 fra en CO-inneholdende gasstrøm ved en forbedret kontinuerlig katalytisk skift reaksjon ved høyt trykk. Reaksjonen finner sted i en eller flere skift-reaktorer ved et superatmosfærisk trykk

på fra 35-250 atm. og en temperatur på mellom 287°C– 537°C. Patentskriftet beskriver ikke reformering av naturgass.

Fra EP 0 000 993-A1 er det kjent en fremgangsmåte for å fremstille ammoniakk ved en primær og en sekundær katalytisk reformering av en hydrokarbonstrøm ved superatmosfærisk trykk. Fra den primære katalytiske reformeringen er forholdet av damp til karbon fra 2,5-3,5, trykket er fra 30 – 120 bar og temperaturen ut av reaktoren er fra 750-850°C. Fra den sekundære katalytiske reformeringen er innholdet av metan fra 0,2 – 10 vekt% på tørr basis og forholdet av hydrogen til nitrogen er fra 2,2 – 2,7. Den sekundære reformeringen tilsettes et overskudd av luft for fremstilling av en gass med et høyere innhold av metan, dvs. ved en lavere temperatur, og/eller et lavere dampforhold og/eller et høyere trykk . I det ovennevnte EP patentet fjernes CO_2 ved lavt trykk ved at hydrogen tas ut ved forhøyet trykk for videre anvendelse ved fremstilling av ammoniakk.

EP 0 289 419 beskriver katalytisk dampreformering av hydrokarboner for fremstilling av hydrogen i en ammoniakkprosess. Den katalytiske dampreformeringen finner sted ved et trykk fra 25 – 120 bar, en temperatur fra 800-1000°C og ved et forhold av damp:karbon på 1,8-2,5. Prosessen drives slik at det er mindre enn 0,3% forurensninger i den H_2-rike gassen som skal benyttes til ammoniakkproduksjon. Foreliggende oppfinnelse tillater et høyere innhold enn 0,3% av CO, CO_2 og CH_4 i den H_2-rike gasstrømmen.

CA 868,821 beskriver fremstilling av syntesegass ved dampreformering av hydrokarboner i en gass og væske ved 50-250 abs. atm., fortrinnsvis 160 abs. atm. for produksjon av ammoniakk og metanol.

Kjent teknikk omhandler ikke en ett-trinns prosess for produksjon av CO_2 -rik gass og H_2 -rik gass under superkritiske betingelser for vann, hvor CO_2 -rik gassblanding tas ut ved et forhøyet trykk i intervallet fra 20 til 200 bar for injeksjon eller deponering i marine formasjoner. Foreliggende oppfinnelse medfører

reduserte kompresjonskostnader ved deponering eller injisering i marine formasjoner som følge av den CO_2 -rike gassblanding tas ut ved et forhøyet trykk.

Foreliggende oppfinnelse omfatter en fremgangsmåte for fremstilling av en CO_2-rik gasstrøm og en hydrogenrik gasstrøm, hvor fremgangsmåten omfatter følgende trinn:

a. naturgass og H_2O fødes til en ett-trinns reformeringsprosess for fremstilling av en gassblanding omfattende CO_2 og H_2 under superkritiske betingelser for vann;
b. gassblandingen fra a) separeres i henholdsvis en H_2-rik og en CO_2-rik gasstrøm.

Videre er temperaturen i reformeringsreaktoren fra ca. 400°C til ca. 600°C, og fremgangsmåten er også kjennetegnet ved et trykk i reformeringsreaktoren fra ca. 200 til ca. 500 bar. Den CO_2 -rike gasstrømmen fra separasjonsenheten foreligger ved et trykk i intervallet fra 20 til 200 bar. I foreliggende oppfinnelse kan blandingen i reformeringsreaktoren føres over et katalysatorsjikt. Reformeringen kan også utføres uten katalysator. Foreliggende oppfinnelse omhandler også anvendelse av den CO_2-rike gasstrømmen fremstilt ifølge den tidligere omtalte fremgangsmåten hvor den CO_2-rike gasstrømmen injiseres i marine formasjoner. Videre omfatter oppfinnelsen anvendelse av den H_2 -rik gasstrøm fremstilt ifølge oppfinnelsen hvor den H_2 -rik gasstrømmen kan nyttegjøres ved hydrogenering, i elektrisitetsproduksjon og som energikilde/drivstoff i brenselceller.

Følgende reaksjon finner sted under reformeringen:

$$CH_4 + 2H_2O \rightarrow CO_2 + 4H_2$$

Reformeringsreaktoren drives ved superkritiske betingelser for vann. Temperaturen i reformeringsreaktoren er fra ca. 400°C til ca. 600°C og trykket i reformeringsreaktoren er fra ca. 200 til ca. 500 bar. Det er et formål ved foreliggende oppfinnelse at CO_2 separeres fra gasstømmen ved et trykk på minst

20 bar og maksimalt på 200 bar før injeksjon i marine formasjoner eller ved deponering. Reformeringsreaksjonen finner sted over et egnet katalysatorsjikt. Reformeringen kan også finne sted uten katalysator i reformeringsreaktoren. Det er også et formål ved foreliggende oppfinnelse å anvende fremstilt H_2 ifølge oppfinnelsens fremgangsmåte for hydrogenering, samt til elektrisistetsproduksjon. Videre er anvendelse av H_2 som energikilde/drivstoff i brenselceller også omfattet av foreliggende oppfinnelse.

CO_2 er en sur gass, og den mest brukte fremgangsmåten for å separere nevnte gass fra andre ikke-sure gassmolekyler er absorpsjon. Ved absorpsjon utnyttes de ulike kjemiske egenskapene til gassmolekylene. Ved å bringe gassblandingen i kontakt med en basisk væske vil de sure gassene i stor utstrekning løses i væsken . Væsken skilles fra gassen og den absorberte gassen kan så frigjøres enten ved å endre væskens sammensetning eller endre trykk og temperatur. For separasjon av CO_2 er det i hovedsak vandige løsninger av alkoholaminer som benyttes. Absorpsjonen skjer ved relativt lav temperatur og høyt trykk, mens stripping av gassen fra væsken skjer ved relativt høy temperatur og lavt trykk. Ved frigjøring av CO_2 fra amin fasen i strippeenheten anvendes vanligvis strippedamp. Dersom partialtrykket av CO_2 i gassen inn på absorber er høyt f.eks. høyere enn 15 bar er det mulig å oppnå høye konsentrasjoner i aminfasen, og en stor del av absorbert CO_2 kan frigjøres i strippeenheten ved forhøyet trykk f.eks. 5-8 bar.

Ved bruk av en eller flere semipermeable membranenheter kan man oppnå at molekyler med ulik molvekt og ulike kjemiske egenskaper permeerer membranen med ulik hastighet. Dette prinsippet kan benyttes for å separere gasser. For den aktuelle gassblandingen kan man velge membraner der H_2 permeerer raskt mens CO_2 permeerer langsomt, hvoretter man oppnår en delvis separasjon av de ulike gasskomponentene. Ved å kombinere fast-stoff membraner og væskemembraner er det også mulig å oppnå at CO_2 permeerer raskt mens H_2 holdes tilbake. Ved ulike separasjonsmetoder kan det være vanskelig å oppnå fullstendig separasjon av de ulike gasskomponentene. Dette er spesielt tilfelle ved bruk av membraner.

For gassblandinger som skal brennes vil en delvis separasjon av hydrogen og CO_2 være tilstrekkelig.

Ved foreliggende oppfinnelse er det ønskelig å deponere fraseparert CO_2. Store mengder CO_2 kan deponeres etter flere metoder der de tre mest aktuelle er deponering på store havdyp, deponering i dype vannreservoarer og deponering i oljereservoarer der gassen samtidig fungerer som drivmiddel for økt oljeutvinning. De to siste lagringsformer opereres kommersielt. I disse lagringsformer må CO_2 gassen bringes opp til høyt trykk for transport i rørledninger frem til deponeringsbrønn og videre til injisering. Injeksjonstrykket vil variere, men vil kunne ligge i området 50 til 300 bar. Dersom CO_2 gassen kan separeres fra H_2/CO_2 blandingen ved forhøyet trykk, kan betydelig kompresjonsarbeid spares noe som er tilfelle i foreliggende oppfinnelse.

Oppfinnelsen belyses nærmere i figur 1. Naturgass (1) føres fra et olje-/gassfelt, blandes med H_2O(2) før blandingen føres til reformering ved superkritiske betingelser. Fremstilt syntesegass (3) separeres ved høyt trykk i to strømmer, henholdsvis en CO_2-rik strøm (5) som reinjiseres i et olje-/gassfelt og en H_2 -rik strøm (4). Den H_2 -rike strømmen anvendes videre til hydrogenering, som energikilde i brenselceller samt ved elektristetesproduksjon.

Eksempel 1:

Eksempelet vedrører en eller flere membranenheter hvor den CO_2-rike gassen kan ha et trykk omtrent lik partialtrykket av CO_2 inn på separasjonsenheten noe som er vist i tabell 1 nedenfor.

Tabell1

Det totale innløpstrykket på separasjonsenheten (bar)	Partialtrykket av CO2 ut fra separasjonsenheten (bar)
200	40
250	50
300	60

Eksempel 2:

Superkritiske betingelser oppstår ved trykk på over 220 bar og temperaturer over 374°C.

I dette eksempelet beskrives ved hvilke betingelser superkritiske forhold oppstår i foreliggende reaktor.

Sammenhengen mellom temperatur og trykk i reaktoren i foreliggende oppfinnelse er som vist i tabell 2. Superkritiske forhold oppstår i reaktoren når verdiene for trykk og temperatur er høyere enn vist i tabell 2.

Tabell2

Trykk (bar)	Temperatur (°C)
1100	354
750	356
500	362
300	368
220	374

Sammenheng mellom temperatur og % fraksjon H_20 ved 220 bar er vist i tabell 3. Superkritiske forhold oppstår i reaktoren når verdiene for temperatur og % fraksjon H_20 ved et trykk på 220 bar er høyere enn vist i tabell 3.

Tabell 3

% fraksjon H_2O (P =220bar)	Temperatur (°C)
0,95	372
0,85	365
0,75	353

Sammenhengen mellom trykk og % fraksjon H_20 ved en temperatur på 374°C er vist i tabell 4. Superkritiske forhold oppstår i reaktoren når verdiene for trykk og % fraksjon H_20 ved 374°C er høyere enn vist i tabell 4.

Tabell 4

% fraksjon H_2O (T= 374°C)	Trykk (bar)
0,95	300
0,85	400
0,75	1000

Patentkrav

1. Fremgangsmåte for fremstilling av en CO_2-rik gasstrøm til injeksjonsformål eller deponering, og en hydrogenrik gasstrøm, karakterisert ved at fremgangsmåten omfatter følgende trinn:
   a. naturgass og H_2O fødes til en ett-trinns reformeringsprosess for fremstilling av en gassblanding omfattende CO_2 og H_2 under superkritiske betingelser for vann;
   b. gassblandingen fra a) separeres i henholdsvis en H_2-rik og en CO_2-rik gasstrøm.

2. Fremgangsmåte ifølge krav 1, karakterisert ved at temperaturen i reformeringsreaktoren er fra ca. 400°C til ca. 600°C.

3. Fremgangsmåte ifølge kravene 1-2,karakterisert ved at trykket i reformeringsreaktoren er fra ca. 200 til ca. 500 bar.

4. Fremgangsmåte ifølge kravene 1-3, karakterisert ved at CO_2 -rike gasstrømmen foreligger ved et trykk i intervallet fra 20 til 200 bar.

5. Fremgangsmåte ifølge kravene 1-4, karakterisert ved at blandingen i reformeringsreaktoren føres over et katalysatorsjikt.

6. Fremgangsmåte ifølge kravene 1-5, karakterisert ved at reaksjonen i reformeringsreaktoren utføres uten katalysator.

7. Anvendelse av CO_2-rik gasstrøm ifølge krav 1 for injeksjon i marine formasjoner.

8. Anvendelse av H_2 -rik gasstrøm fremstilt ifølge krav 1 for hydrogenering.

9. Anvendelse av H_2 -rik gasstrøm fremstilt ifølge krav 1 som energikilde/drivstoff i brenselceller.

10. Anvendelse av H_2 -rik gasstrøm fremstilt ifølge krav 1 for elektrisitetsproduksjon.

Sammendrag

Foreliggende oppfinnelse vedrører en fremgangsmåte for fremstilling av en CO_2-rik gasstrøm til injeksjonsformål eller deponering, og en hydrogenrik gasstrøm, hvor fremgangsmåten omfatter følgende trinn:

a. naturgass og H_2O fødes til en ett-trinns reformeringsprosess for fremstilling av en gassblanding omfattende CO_2 og H_2 under superkritiske betingelser for vann fra ca. 400°C til ca. 600°C, og trykket fra ca. 200 til ca. 500 bar i reformeringsreaktoren;

b. gassblandingen fra a) separeres i henholdsvis en H_2-rik og en CO_2-rik gasstrøm.

Anvendelse av CO_2-rik gasstrøm for injeksjon i marine formasjoner samt anvendelse av H_2 -rik gasstrøm for hydrogenering, som energikilde/drivstoff i brenselceller og for elektrisitetsproduksjon er også omfattet av oppfinnelsen.